HIGH-TEMPERATURE SYSTEMS FOR NUCLEAR PROCESS HEAT

By James P. McGee

* * * * * * * * * * * * information circular 7954

UNITED STATES DEPARTMENT OF THE INTERIOR
Fred A. Seaton, Secretary

BUREAU OF MINES
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by

James P. McGee[2/]

SUMMARY

Use of nuclear reactors to furnish high-temperature heat is a field that has not received as much attention as propulsion and electric power production. Investigations are being conducted under a joint Atomic Energy Commission (AEC)--Bureau of Mines research program to determine the feasibility of using nuclear heat for chemical processing.

A part of this process-heat-reactor program is the study of a system in which helium transfers heat from a nuclear reactor to a chemical processing vessel. This requires construction of (1) a nuclear reactor to operate with a helium exit temperature of 2,500° F. and with pressures in the range of 250 to 500 p.s.i.g. and (2) a processing vessel (heat exchanger) to convert the enthalpy of the helium into chemical process reaction heat. Neither component exists at present, and development of each promises to be a formidable task.

To aid in the study of such a system, the Bureau of Mines has designed and constructed a vessel heated by electrical induction to simulate a source of nuclear fission heat. This vessel is undergoing preliminary testing.

Gasification of coal and reforming of hydrocarbons are promising uses for nuclear heat. It appears that a higher price per unit of nuclear heat can be tolerated for chemical processing than that allowable for power generation.

INTRODUCTION

Although nuclear reactors have been constructed for research and testing, electric power production, and propulsion, direct utilization of nuclear heat for chemical processing has not been developed. However, the use of nuclear heat for that purpose could be attractive both technically and economically; AEC, the Bureau of Mines, and major industrial companies have considered this source of heat.

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There are several advantages in using energy produced by nuclear fission for supplying chemical process heat. Temperatures that can be achieved are limited only by materials of construction, since there is no inherent limiting temperature, as in the combustion of hydrocarbon fuels. Nuclear heat can be produced at elevated pressures, and there is no combustion air to compress. By selecting a suitable process cycle, system heat losses can be made negligible.

Nuclear-process heat reactors fit into three general classes, shown in figure 1:

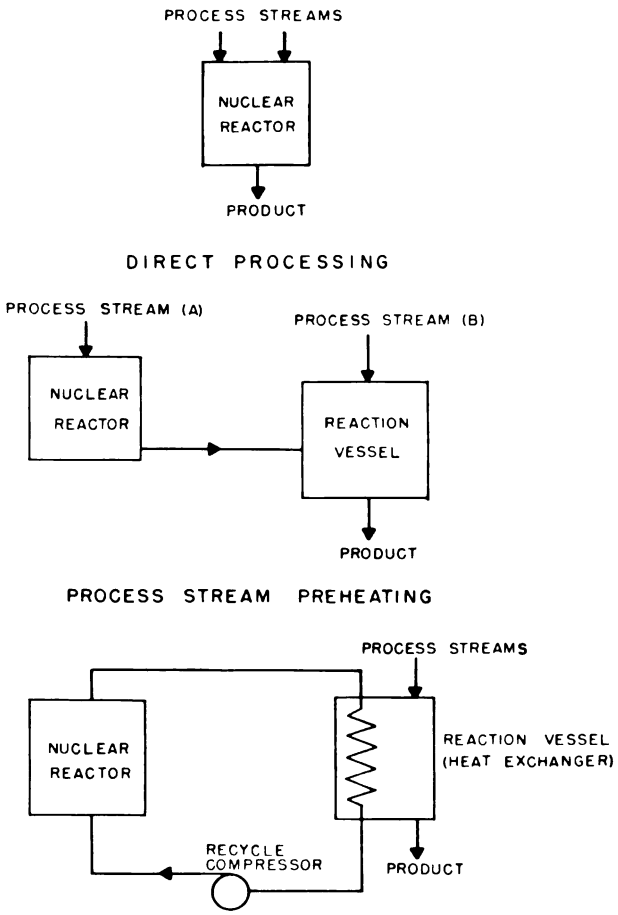

FIGURE 1. - Types of Process Heat Reactors.

1. Direct processing.--All of the process streams passed through and reacted within the nuclear reactor.

2. Process-stream preheating.--One or more of the process streams preheated in the nuclear reactor and subsequently reacted by direct contact in a separate reaction vessel.

3. Indirect process cycle.--An inert gas such as helium heated in the nuclear reactor and subsequently giving up its heat to the process streams by indirect heat exchange in a separate reaction vessel.

These possible types of nuclear reactors have been described by Graham (2).3/ Each type has individual advantages. If satisfactory ceramic fuel elements, capable of containing the fission products can be developed, the direct cycle reactor should be attractive for processes that are favored by neutron and gamma irradiations and where induced radioactivity in the product stream is not a problem. Also, if suitable ceramic fuel elements and refractories are developed, preheating of process streams, such as air or steam, to 3,000° F. and above should solve many industrial requirements for process heat. However, it appears that considerable research and development will be required to perfect such nuclear-fuel elements. Furthermore, a great many operating problems need to be solved, owing to the unprecedented temperature levels involved.

3/ Underlined numbers in parentheses refer to items in the bibliography at the end of this report.

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Operation of a reactor in this temperature range would represent a considerable advance in reactor technology.

ACKNOWLEDGMENTS

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PROCESS HEAT REACTOR SYSTEM

It would seem then that the first approach to the use of high-temperature nuclear heat for processing should be to devise a system that imposes the least restrictions on the nuclear-reactor design.

The use of a gas as a heat-transfer medium has three fundamental disadvantages: (a) The overall heat-transfer coefficients are considerably lower for gases than for liquids; (b) to remove a given amount of heat, the energy required to recirculate a gas is usually higher than that required for a liquid; and (c) the reactor volume will be greater when a gaseous coolant is used as the heat-transfer medium. Gaseous coolants, however, offer the advantage of high-temperature operation at moderate pressures. Gases such as helium are inert chemically, and corrosion problems are virtually eliminated. Gases have been used for a number of years as heat-transfer mediums at temperatures up to 4,000° F. (1).

A system using the indirect cycle will consist essentially of a nuclear reactor to heat the gaseous heat-transfer medium, a high-temperature heat exchanger to transfer heat from the gas to the process streams, and a recycle compressor to recirculate the carrier gas (fig. 2).

Choice of Inert Gas

The criteria for choosing the gaseous heat-transfer medium are:

1. Heat-transfer properties
2. Pumping power requirements
3. Nuclear properties
4. Chemical properties
5. Cost
6. Availability
7. Purity.

Properties of the various gases are shown in table 1. Helium meets the requirements better than any other gas. Its cross section for neutron absorption is negligible. It has a high specific heat and low-pumping power requirement and is inert chemically. The specific heat of hydrogen is more than double that of helium, and the power requirements would be the lowest for any gas. However, from a chemical standpoint hydrogen is not acceptable, since it would react with a graphite moderator or reflector in the 1,000° to 2,500° F. range. Other gases considered were nitrogen, carbon dioxide, and argon. Carbon dioxide also reacts with graphite in the range 1,000° to 2,500° F. The

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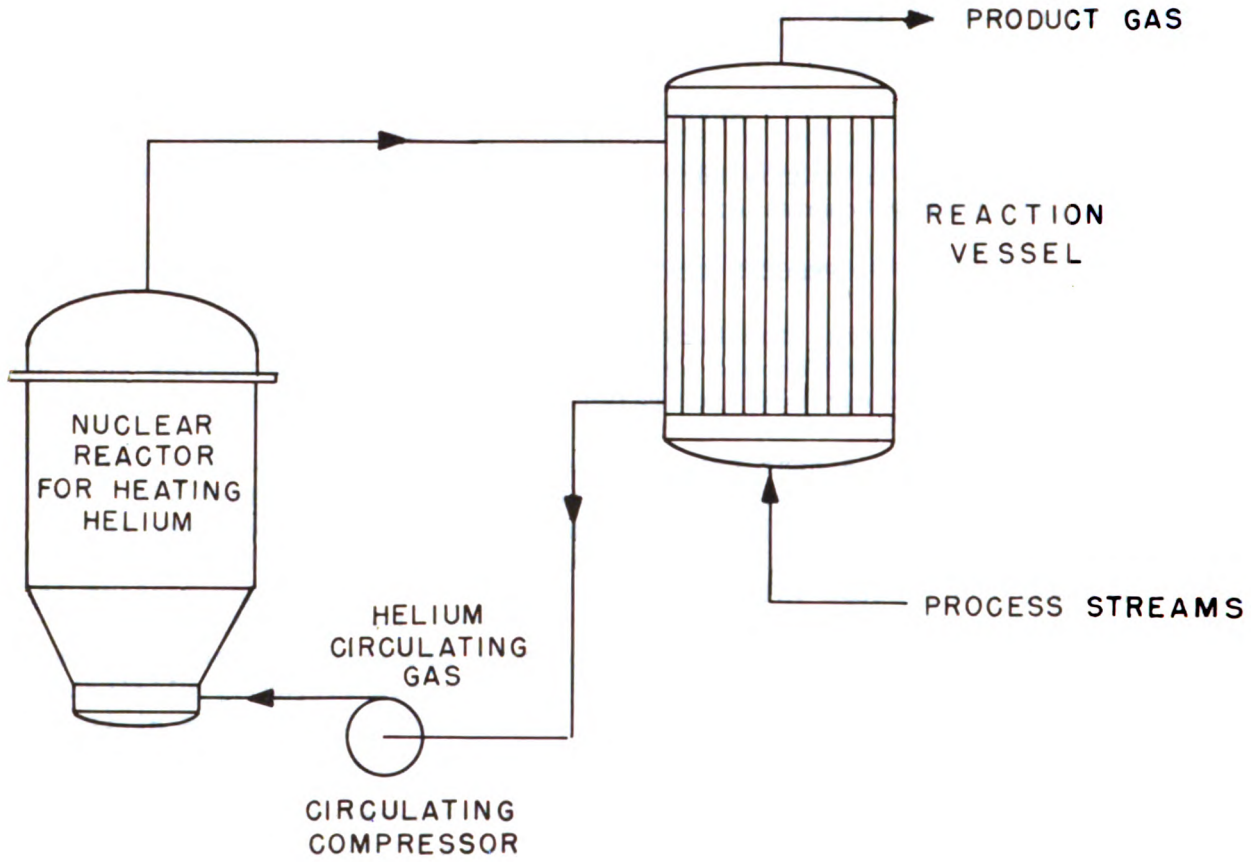

FIGURE 2. - Simplified Helium-Recycle Flowsheet.

use of these gases would preclude unclad graphite as a moderator in the reactor. Argon has a low specific heat which makes pumping costs prohibitive. It also forms the radioactive isotope, A^{41}, which would be difficult to separate. When irradiated, nitrogen forms the radioactive isotope, C^{14}, which has a long half-life. Unfortunately, nitrogen also reacts with ferrous metals at high temperatures, forming nitrides, and has a relatively low specific heat.

Helium has the disadvantage of being expensive and limited in supply. Most of the helium now being produced comes from natural gas in the vicinity of Amarillo, Tex. Current helium production by the Bureau of Mines is about 330,000,000 cubic feet annually. To obtain this helium, approximately 2.5 percent of the helium-bearing natural gas now going to market is being processed. The helium in the other 97.5 percent of the natural gas not processed is lost. Further demands for helium would require more plant capacity for processing additional quantities of natural gas. The U. S. Department of the Interior proposes a conservation program under which 3 billion cubic feet of helium per year would be recovered. Any gas not immediately needed would be stored underground.

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TABLE 1. - Properties of gaseous coolants

Gas	Molecular weight	Specific volume, cu.ft./lb.	Specific heat, Cp B.t.u./lb.°F. at 2,500°F.	Thermal absorption cross section, barns	Nuclear properties	Chemical properties
Argon	40	9.5	0.124	0.62	Forms radioactive argon 41	Inert.
Carbon dioxide	44	8.54	.336	.003		Reacts with graphite at high temp.
Helium	4	95.2	1.24	.007	Inert	Inert.
Hydrogen	2	187.9	3.94	.32		Reacts with graphite at high pressures. As temp. increases, extent of reaction decreases. Reacts with uranium compounds.
Nitrogen	28	13.5	.291	1.7	Forms radioactive carbide 14	Reacts with ferrous metals at high temp.

The quantity of helium required by a reactor system probably will be in the range of 1 cubic foot of gas per thermal kilowatt of capacity. Due to the high cost of helium, it is obvious that gas-cooled reactors using helium must be virtually gastight, even if the fuel element retains the fission products. Also, provisions must be made for draining the system and storing the helium during refueling and maintenance.

BUREAU OF MINES EXPERIMENTAL PROGRAM

The Bureau is interested primarily in the application of the nuclear heat to the gasification of coal and the AEC in the design of the nuclear reactor to furnish this heat. A nonnuclear system was designed and built by the Bureau of Mines as part of the cooperative process-heat reactor program, to serve as a prototype for a nuclear process heat system.

In this system electric induction heat is used to simulate nuclear fission. The heat-transfer fluid is helium, which, in turn, gives up its energy by indirect transfer to air at high temperatures (fig. 3). Helium is heated to 2,500° F. in the induction simulated reactor (ISR). The temperature is then reduced to 1,000° F. by indirect exchange with air in the shell and tube

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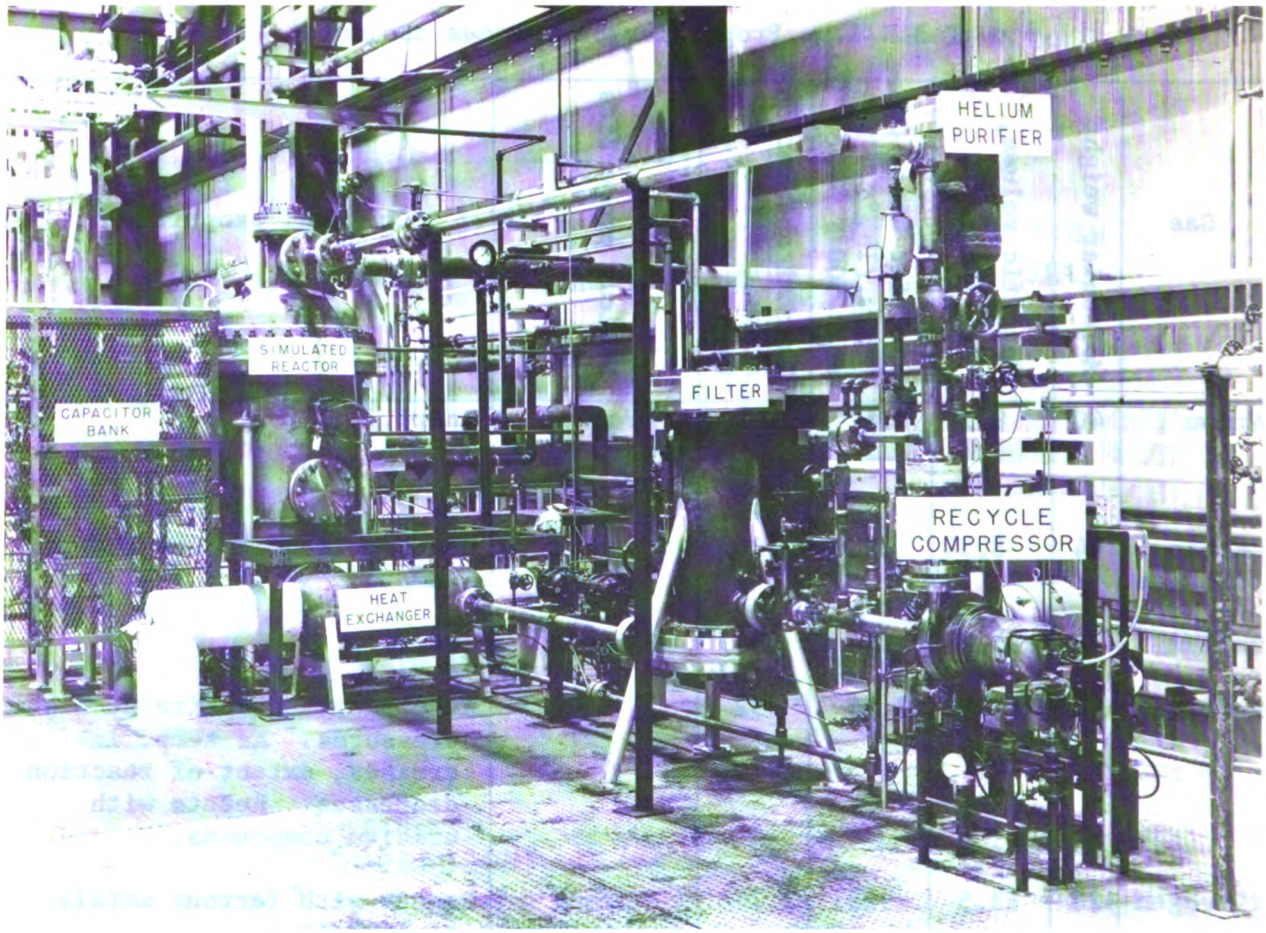

FIGURE 3. - 2,500° F., 250 p.s.i.g. Helium Recycle Loop.

heat exchanger. The helium at 1,000° F. is filtered to remove particles that could damage the high-speed recycle compressor. This compressor provides the motive force to recycle the helium back to the ISR.

Air is recycled through the heat exchanger to provide the cooling medium for the helium. Standard equipment is used in the air circuit and includes a reciprocating recycle compressor, air precooler, oil separator, and an electric preheater. The preheater is installed for eventual use in the investigation of high-temperature heat exchangers. In the helium circuit the induction heated reactor, helium-to-air heat exchanger, and the recycle compressor are development items and will be described in detail.

1. Induction simulated reactor (ISR).--The induction simulated reactor is a type 304 stainless steel pressure vessel. It is designed for 300 p.s.i.g. with an operating pressure of 250 p.s.i.g. A sectional view of the construction is shown in figure 4. The vessel functions as the source for transferring heat to the recycled helium. Heating is accomplished by a 6-inch-i.d. by 24-inch-long bed of graphite spheres installed inside an induction coil. To minimize heat loss, two ceramic tubes and a layer of granular insulation are

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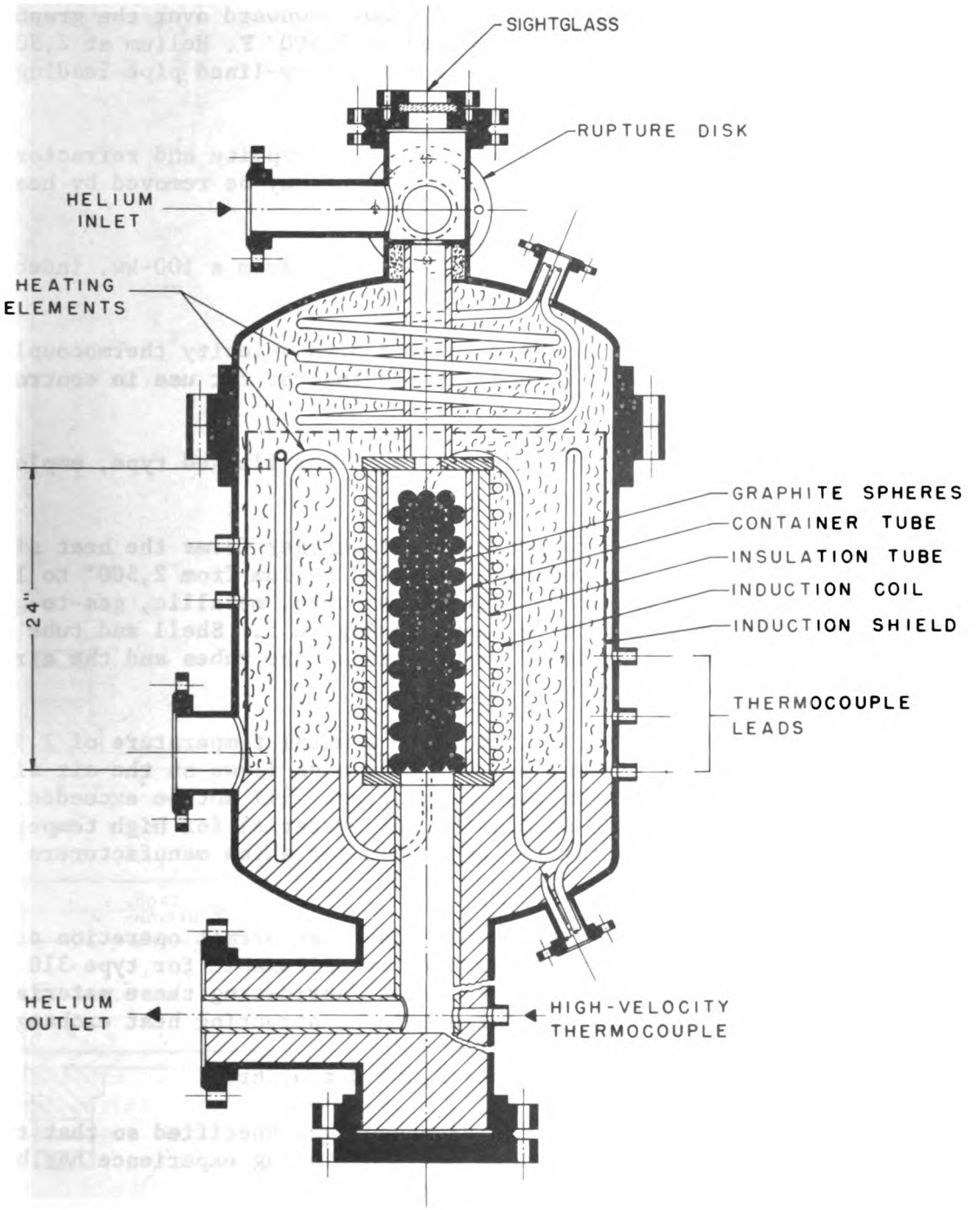

FIGURE 4. - 250 p.s.i.g. Induction Simulated Reactor (ISR).

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installed between the heating elements and the induction coil. Helium at 1,000° F. enters the top of the vessel and flows downward over the graphite elements, where the gas temperature is raised to 2,500° F. Helium at 2,500° F. leaves the bottom of the reactor through a refractory-lined pipe leading to the heat exchanger.

Auxiliary heating coils are installed in the graphite and refractory insulation so that occluded gas and moisture present may be removed by heating while the vessel is evacuated.

The energy for the induction field is obtained from a 100-kw. induction generator operating at 9,600 cycles per second.

The exit-gas temperature is measured by a high-velocity thermocouple. Shielded couples are positioned throughout the reactor for use in controlling the operation.

All of the connections to the vessel are of the flanged type, employing octagonal ring joint seals.

2. Helium-to-air heat exchanger.--This exchanger forms the heat sink for the system. Its function is to cool the recycled helium from 2,500° to 1,000° F. It also serves as a prototype for a high-temperature, metallic, gas-to-gas heat exchanger. The construction is shown in figure 5. Shell and tube construction is used, with the helium flowing outside the tubes and the air circulating through the tubes.

The exchanger, as installed, is limited to a metal temperature of 1,900° F., with balanced pressures of air and helium. The mass flows on the air side of the tube have been so chosen that this temperature will not be exceeded. The tubes are of type 310 stainless steel, the best material for high temperature and oxidation resistance commercially available for which manufacturers welding techniques are fully developed.

Metals and alloys currently being developed may permit operation at several hundred degrees higher than the temperature allowable for type 310 stainless steel. When more experience is gained in fabricating these materials, they will be tested for suitability for use in constructing heat exchangers.

The heat load on the exchanger is 112,000 B.t.u./hr.

A low helium velocity through the exchanger was specified so that the system pressure drop could be minimized until operating experience has been gained with the centrifugal recycle compressor.

The helium mass flow "G" (lb./hr./ft.^{2}) through the exchanger is approximately 1,000, and the overall heat-transfer coefficient "U" (B.t.u./hr./sq. ft./°F.) was calculated to be 14. The heating surface based on the outside diameter of the tubes is 12.6 sq. ft.

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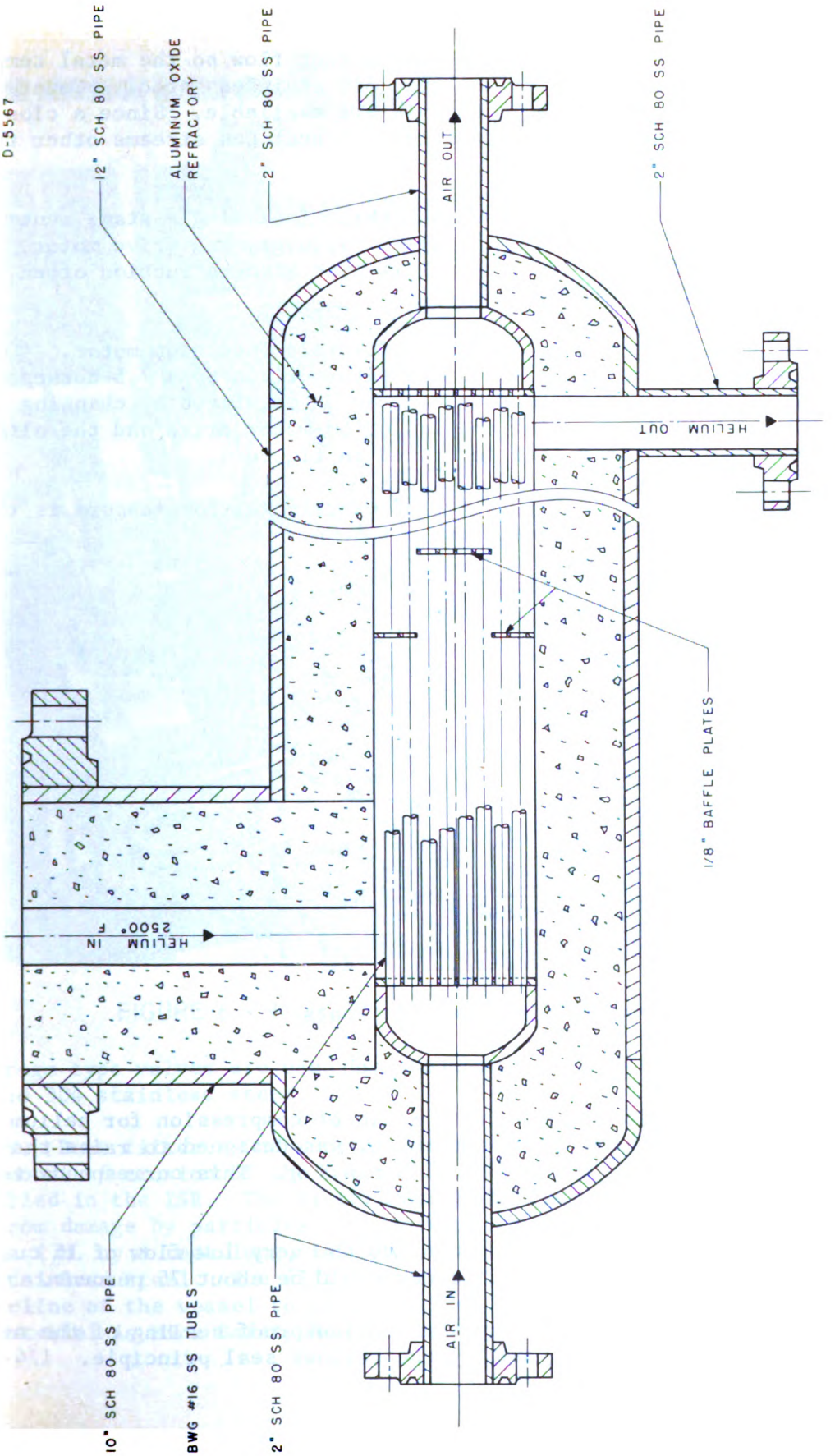

FIGURE 5. - Helium-to-Air Heat Exchanger.

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The exchanger is arranged for parallel-current flow so the metal temperatures will not exceed the limits of the type 310 stainless steel. Countercurrent flow will be used when better materials are available. Since a closed circuit has been adopted, it will be possible to heat gas streams other than air.

3. Helium-recycle compressor.--This machine is a single-stage centrifugal type, consisting of an overhung impeller, diffuser, and drive motor. The entire assembly is hermetically sealed. This type of construction often is referred to as "canning."

The drive is a 2-horsepower, 3-phase, 400-cycle induction motor. 400-cycle current is obtained from a 5-kw. alternator driven by a 7.5-horsepower, 60-cycle induction motor. Compressor speed can be regulated by changing the pulley size of the V-belts coupling the induction-motor drive and the alternator. The recycle compressor is illustrated in figure 6.

An important parameter in the design of centrifugal compressors is the adiabatic head against which the machine must pump.

Adiabatic head may be expressed in the form,

$$Ha = R\, T_1 \, \frac{k}{k-1} \left[(r)^{\left(\frac{k-1}{k}\right)} - 1 \right]$$

Ha = adiabatic head - ft.

$R = \frac{1544}{\text{mol weight}}$ - ft./°R.

T_1 = gas temperature - °R.

k = ratio of specific heats

r = ratio of compression

The variation of adiabatic head with ratio of compression for helium at 1,200° F. is shown in figure 7. The compressor was designed to raise the pressure of 1,000° F. helium from 260 to 265 p.s.i.g. This corresponds to an adiabatic head of approximately 11,000 ft.

Low efficiency is to be expected owing to the very low flow of 15 cu.ft./min. In a commercial machine the efficiency would be about 75 percent.

4. Valves.--All valves in the system use leakproof sealing of the stems. The larger valves (1- and 2-inch) employ the bellows seal principle. 1/4-inch

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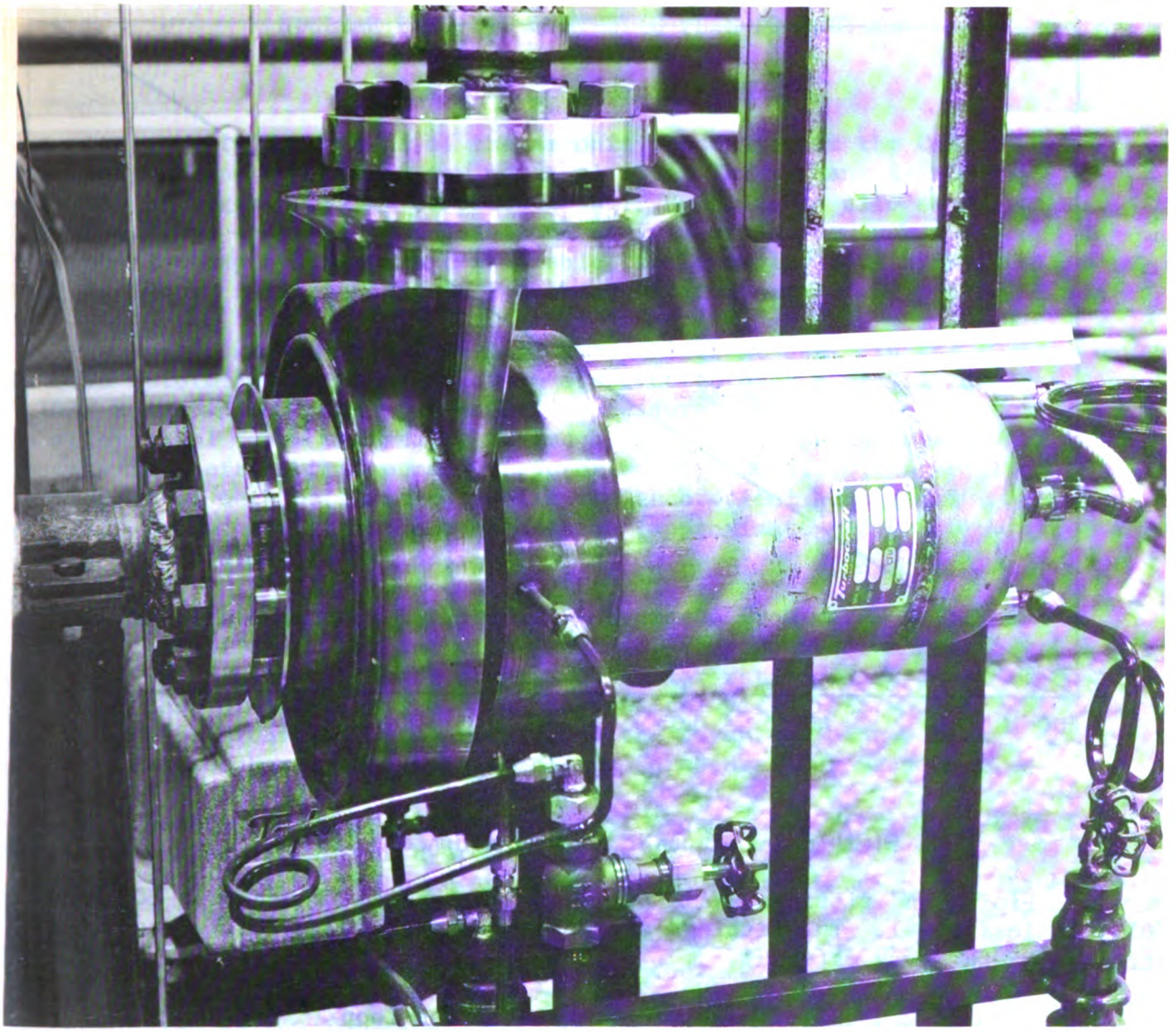

FIGURE 6. - Recycle Compressor for High-Temperature Helium.

diaphragm-type valves are used for instrumentation. All valves are constructed of type 304 stainless steel.

5. Dust filter.--One function of the dust filter is to aid in determining the amount of erosion of the ceramic materials and graphite heating elements installed in the ISR. The filter also protects the high-speed recycle compressor from damage by particles entrained in the helium. The filter vessel is 12 inches i.d. by 3 feet long, flanged at top and bottom, and constructed of type 304 stainless steel. The filter element is arranged longitudinally across the centerline of the vessel to give a filter surface 1 foot by 3 feet. The filter material is Mine Safety Appliance No. 970-F Fiberglass.

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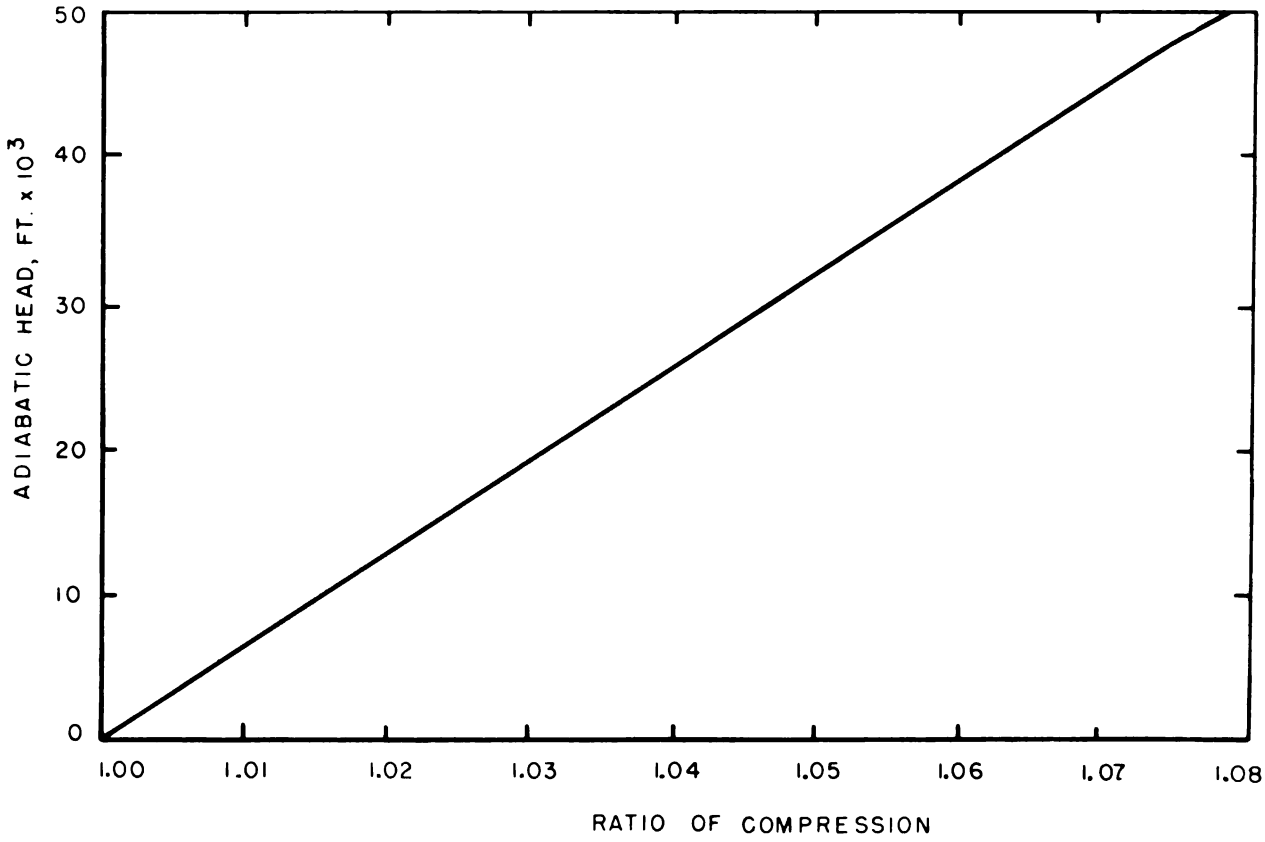

FIGURE 7. - Adiabatic Head for Helium at 1,200° F.

6. Piping and fittings.--All piping in the system is schedule 80, type 304 stainless steel. The fittings are designed for 2,000 p.s.i.g. and constructed of the same material.

Preliminary Operation of the Loop

The loop has been operated for short periods above 2,200° F. The major cause for shutdowns has been the recycle compressor. Warpage of the shaft has caused the impeller to rub the casing. A new impeller and shaft design will use Discaloy for the impeller and type 347 stainless steel for the shaft.

An investigation of a gas lubricated, "canned" compressor is continuing.

Another major difficulty in the first series of runs was attack of the graphite spheres by entrained moisture in the system. This trend has been almost eliminated by continuously freezing out the entrained water vapor from a side stream of the recirculated helium. This is done while slowly raising the temperature of the spherical heating elements.

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NUCLEAR HEAT FOR SYNTHETIC FUEL PRODUCTION

Reforming Methane to Hydrogen

At present, 80 percent of the ammonia production in the United States utilizes methane as a basic raw material (3). The methane is reformed to make the hydrogen required to produce NH_3. Hydrogen is also a raw material for many other chemical processes, and many important new uses could be developed if cheap hydrogen were available, for example, coal hydrogenation to produce liquid fuels and direct reduction of iron ore.

Two processes to reform methane are in current use. In the first, known as tube reforming, methane and steam are reacted over a nickel catalyst at 1,500° F. at approximately atmospheric pressure. The catalyst is suspended in alloy steel tubes externally heated by combustion of methane and air. The process is inefficient thermally; approximately 700 B.t.u. is required per cubic foot of methane reformed, while the theoretical heat requirement is only 220 B.t.u. per cubic foot of methane (approximately 250 B.t.u., including losses from thermal radiation).

An alternate process for producing hydrogen from methane depends upon partial combustion of methane with oxygen of 95-percent or higher purity, followed by water-gas shift with steam. The molar ratio of oxygen to methane is 0.5, and that of steam is 1.0. The theoretical yield is 3 volumes of hydrogen per volume of methane. This process has the advantage of generating the hydrogen at 30 atmospheres, the most economical pressures for many uses.

A schematic diagram of a hypothetical process for reforming methane (or other low-molecular-weight hydrocarbon gases) with steam, using nuclear heat, is shown in figure 8. Methane and steam are passed through a heat exchanger, the tubes of which are filled with a nickel catalyst to promote the reforming of methane to form hydrogen and carbon monoxide. The reaction is carried out at 1,700° F. and 30 atmospheres and is endothermic. Heat for the reaction is supplied by helium heated to 2,000° F. at 30 atmospheres pressure in a nuclear reactor. The helium leaves the heat exchanger at 500° F. and is recycled to the nuclear reactor. Two mols of steam are required for each mol. of methane, and the theoretical yield, after water-gas shift, is 4 volumes of hydrogen per volume of methane.

In comparing the proposed nuclear process with tube reforming, the unique quality of nuclear heat using a recycle gas as the heat carrier is the elimination of stack loss. The unused portion of the heat in the carrier gas is returned to the reactor. The only heat loss is through radiation. Nuclear heat therefore could be about 3 times as efficient as the tube reforming method (250 B.t.u. vs. 700 B.t.u.). If methane costs 35 cents per 1,000 std.c.f. or 35 cents per 10^6 B.t.u., nuclear heat at \$1.00 per 10^6 B.t.u. appears to be competitive. The conventional tube process cannot generate the hydrogen at pressure, since at these temperatures a balanced pressure must be maintained across the tubes to eliminate stress. Compression costs for the methane and air used to supply heat for the reforming make this system uneconomical. In the nuclear process, moreover, the hydrogen is generated

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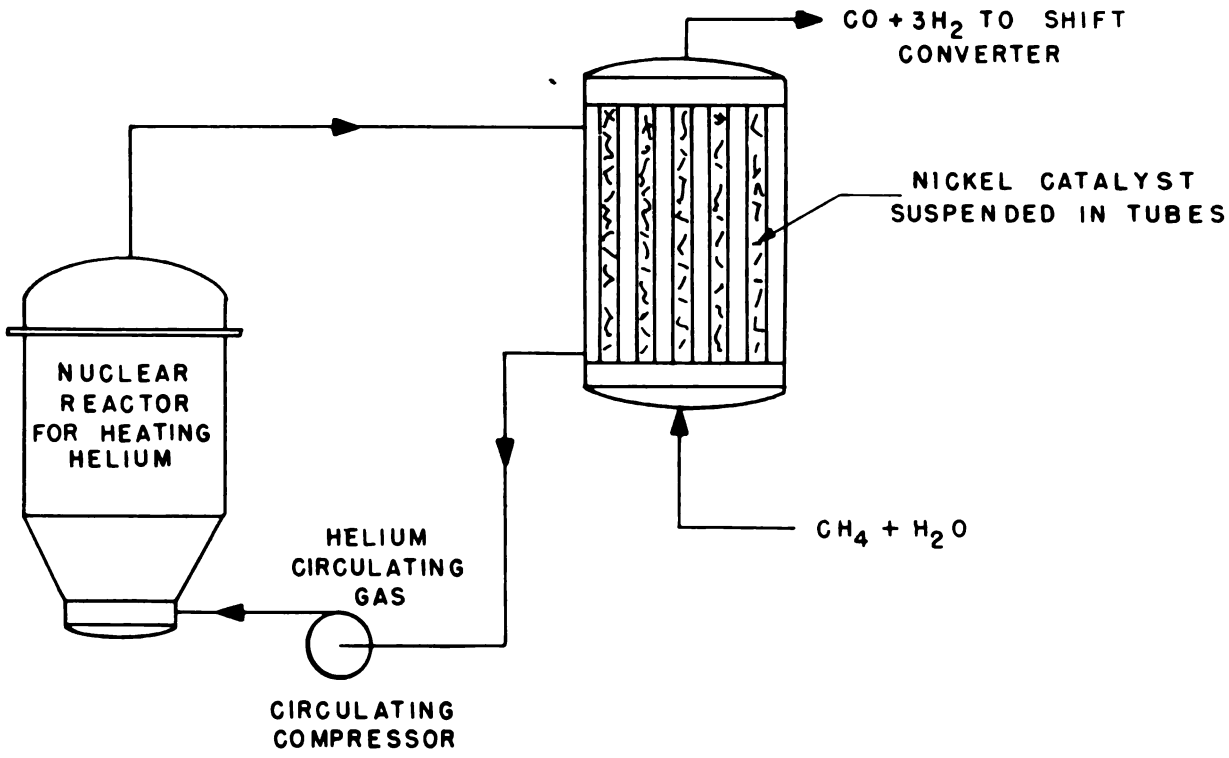
CO + 3H2 TO SHIFT CONVERTER
NICKEL CATALYST SUSPENDED IN TUBES
NUCLEAR REACTOR FOR HEATING HELIUM
HELIUM CIRCULATING GAS
CH4 + H2O
CIRCULATING COMPRESSOR

FIGURE 8. - Simplified Flowsheet for Reforming Methane, Using Nuclear Heat.

at 30 atmospheres pressure, and an additional 8 cents per 1,000 std.c.f. in compression costs is saved if the hydrogen is used at 30 atmospheres or above.

In comparing the nuclear process with the partial oxidation process, 25 percent of the methane per unit volume of hydrogen produced is saved, and the cost of oxygen is eliminated. For 1,000 std.c.f. methane processed, the theoretical oxygen requirements are approximately 500 std.c.f. At a cost of $5 per ton of 95-percent oxygen, the 500 std.c.f. of oxygen has a value of 10.6 cents. The cost of compressing the 500 std.c.f. of oxygen to 30 atmospheres adds another 4 cents.

The potential savings in using nuclear energy are $1.23 for each 4,000 std.c.f. of methane processed (16,000 std.c.f. of hydrogen produced) which requires 10^6 B.t.u. Therefore, nuclear energy would be competitive at $1.23 per 10^6 B.t.u.

TABLE 2. - Comparative costs for hydrogen production

(Basis, 16,000 cu.ft. H_2)

	Nuclear heating		Tube reforming	
	Quantity, cu.ft.	Costs	Quantity, cu.ft.	Costs
Methane.....	4,000	$1.40	5,330	$1.86
O_2 at 30 atm.....	0	0	2,665	.78
Total.....		1.40		2.64

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Coal Gasification

Reserves of coal in the United States and in many other highly industrialized countries are much larger than of petroleum or natural gas. Therefore, at some future date a large part of liquid and gaseous fuels is expected to be produced from synthesis gas derived from coal.

For many years almost all hydrogen and synthesis gas were made by the cyclic water-gas process. Because of the high cost of coke in the United States, alternate methods for gasifying coal have been studied; the most highly developed of these methods is partial combustion of coal with oxygen at elevated pressures. In this process, steam, oxygen, and coal are reacted in a pressure vessel at 30 atmospheres. The quantities of reactants required to produce 1,000 std.c.f. of a carbon monoxide-hydrogen mixture are about 35 pounds of bituminous coal, 300 std.c.f. of 95-percent oxygen, and 12 pounds of steam.

A proposed process using nuclear heat for coal gasification is shown diagrammatically in figure 9. Coal and water in the ratio of approximately 1.5 pounds water to 1 pound of coal (dry ash-free basis) are introduced into a slurry mix tank. After suitable mixing the slurry is pumped through preheater tubes, where the water is flashed into steam and the coal-steam mixture is preheated to 600° F.

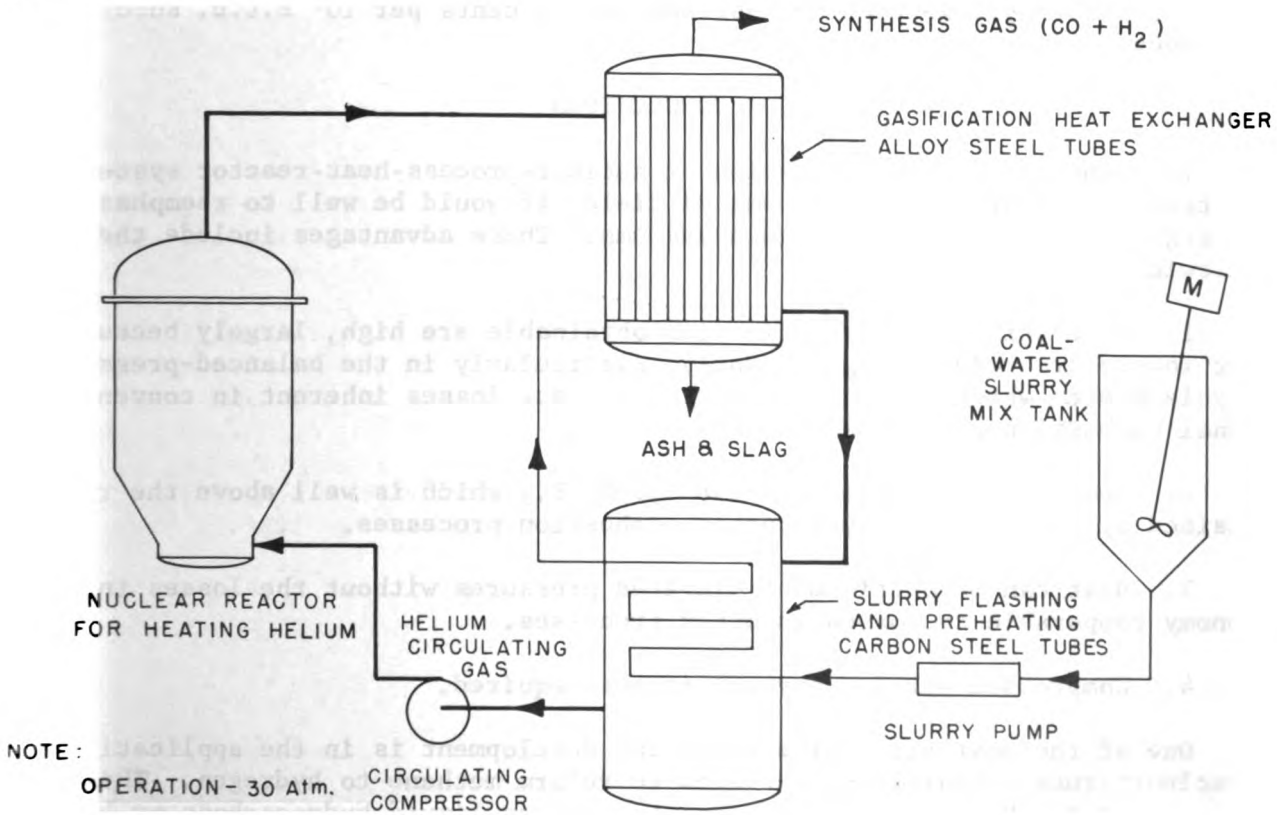

FIGURE 9. - Simplified Flowsheet for Nuclear Gasification.

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The steam-coal mixture then enters the gasifier, where the temperature is raised to the range of 1,700° to 2,500° F. and the heat is supplied for the endothermic steam-carbon reaction. The heating medium for both the preheater and gasifier is helium recycled through a nuclear reactor. The pressures of both the recycled helium and the reactants are balanced at 30 atmospheres. This eliminates pressure stresses in the tubes and allows the tube material to be used at higher temperatures than if a pressure differential existed. For gasification of a low-rank coal, such as lignite, the reaction temperature is low enough to permit the use of high-alloy metal tubes. For gasification of higher rank coals, such as bituminous, which will require higher temperatures, either ceramic or cermet heat-exchanger tubes may be required.

In contrast to the partial oxidation process the nuclear system has the advantage of eliminating compressed process oxygen and reducing coal requirements 40 percent. This system might require only 22 pounds of coal per 1,000 std.c.f. of $CO + H_2$. For coal at $4 per ton, the reduction of 13 pounds of coal results in a saving of 2.6 cents per 1,000 std.c.f. of $CO + H_2$. For oxygen at $5 per ton, the elimination of 300 std.c.f. saves 6.3 cents, and the elimination of oxygen compression saves at least an additional 2.4 cents. The total reduction of costs of raw materials could amount to 11.3 cents per 1,000 std.c.f. of $CO + H_2$.

The nuclear process requires a heat input of 160,000 B.t.u. per 1,000 std.c.f. ($CO + H_2$), so that nuclear heat at 71 cents per 10^6 B.t.u. should be competitive.

CONCLUSIONS

In summarizing this discussion of nuclear-process-heat-reactor systems and their application in the chemical field, it would be well to reemphasize the significant advantages of such systems. These advantages include the following:

1. Heat-utilization efficiencies obtainable are high, largely because heat losses are reduced significantly, particularly in the balanced-pressure recycle system which operates without the stack losses inherent in conventional combustion processes.

2. Heat can be furnished above 3,000° F., which is well above the range attained by conventional hydrocarbon combustion processes.

3. Heat can be obtained at elevated pressures without the losses in economy comparable to losses in other processes.

4. Compressed air (or oxygen) is not required.

One of the most attractive areas for development is in the application of a nuclear-process-heat-reactor system to reform methane to hydrogen. This is of interest for hydrogenation of coal to produce liquid hydrocarbons or high-B.t.u. gas. Hydrogen is also a basic element in many chemical processes. For reforming methane, nuclear energy furnished at $1.00 to $1.23 per 10^6 B.t.u.

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would be competitive with two conventional processes, tube reforming and partial oxidation.

Finally, the prime interest of the Bureau of Mines is to apply nuclear heat for gasifying coal to produce synthesis gas--another source of hydrogen. The Bureau has designed and constructed a nonnuclear system to serve as a prototype, nuclear-process-heat system, and this system will be adapted to the coal-gasification process. A process adapted for coal gasification probably would be competitive with conventional methods if nuclear heat could be furnished at about 71 cents per 10^6 B.t.u. Also, the need for compressed oxygen would be eliminated and coal requirement reduced, perhaps as much as 40 percent.

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